THE JOURNAL and CERAMIC ABSTRACTS

AMERICAN CERAMIC SOCIETY

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MONTHLY PUBLICATION DEVOTED TO THE ARTS AND SCIENCES RELATED TO THE SILICATE INDUSTRIES

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Publication Office: 20th & Northampton Sts., Easton, Pa.
Editorial Office: 2525 N. High St., Columbus, Ohio

Entered as second-class matter at the post office at Easton, Pa., under the act of March 3, 1879. Published monthly.
Subscription fifteen dollars ($15.00) a year. Single numbers $1.50
(Foreign and Canadian postage, 50¢ additional on subscriptions)
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REVIEW OF THE RELATION OF DENSITY AND REFRACTIVE INDEX TO THE COMPOSITION OF GLASS: II*

By Kuan-Han Sun, Hurd W. Safford, and Alexander Silverman

III. Relation Between Composition and Refractive Index

In many series of glasses, refractive index does not vary linearly with composition, and the relationship that does result depends on the type of glass and the composition range studied.

Zschimmer,[57] using data after Abbé and Schott, plotted refractive indices against (*a*) equivalent weights of Li_2O, ZnO, BaO, and PbO per 100 parts of B_2O_3; (*b*) equivalent weights of Na_2O, ZnO, BaO, and PbO per 100 parts of alkali-silicate glass; and (*c*) percentage of B_2O_3 in alkali-silicate glass. In no case was a straight line produced, and there was a curve-maximum in (*c*). The following qualitative conclusions were drawn: (i) The mean refractive indices of B_2O_3, fused silica, and alkali-silicate glass are raised by the solution of metallic oxides in increments not proportional to the molecular weight of the dissolved oxide; (ii) that variation with concentration is not direct is evident because the optical constants decreased for Li_2O (in B_2O_3), Na_2O (in SiO_2), and ZnO (in B_2O_3 and alkali-silicate glass) and increased for PbO and BaO; (iii) B_2O_3 up to 15% in potassium-silicate glass increases refractive index, and further additions reverse the effect.

Peddle[58] plotted refractive index against moles of a variable constituent. For the soda-lime-silica glasses studied, he was able to calculate that composition necessary to produce a definite value of refractive and dispersion index.[59] His graphs did not indicate a straight-line relation between refractive index and composition.

(A) Additive Calculations and Oxide and Compound Factors

Finn and Thomson,[60] from additive considerations, derived constants for the "partial" indices of refraction of silica, soda, and lime (Table XII).

TABLE XII

REFRACTIVE INDEX CONSTANTS AFTER FINN AND THOMSON[60]

	(*a*)	(*b*)	(*c*)
SiO_2	0.03875	0.8063	−10
Na_2O	0.01406	1.0193	0
CaO	0.01332	1.0690	1

These constants are used in equations (29) and (30). Computed indices of refraction for soda-silica and soda-lime-silica glasses varied from observed values by less than 0.001 and 0.005 unit, respectively.

$$n = n'_A + n'_B + n'_C \qquad (29)$$

$$n'_X = a(p_X + c)^b \qquad (30)$$

wherein n = refractive index
X = any constituent.

Turner[61] indicates that refractive indices are of additive character for glasses of the general series $6SiO_2 \cdot\cdot (2-x)Na_2O \cdot xRO$. English and Turner[62] and Knapp[63] observed smooth maxima and minima in the refractive index-composition curves for soda-borosilicate glasses.

For a series of glasses of the general formula $6SiO_2 \cdot\cdot xR_2O \cdot yCaO$ (wherein R_2O is Na_2O or K_2O or both), Takamatsu[64] found that when $(x + y)$ is less than 2.4 moles, the increment in refractive index varies linearly with increase of alkali oxide and lime content. Above 2.4 moles, the curve takes the shape of a hyperbola.

From the data of Zschimmer, Thiens, and Wright for lead glasses, Mees[65] derived equations for the calculation of refractive and dispersive index.

For glasses containing both K_2O and Na_2O,

$$(n-1)\cdot 10^3 = 0.06472 \cdot p^2 - 0.77157 \cdot p + 523.3 \qquad (31)$$

$$\nu^* = 0.00310 \cdot p^2 - 0.74207 \cdot p + 63.47. \qquad (32)$$

For glasses containing K_2O alone,

$$(n-1)\cdot 10^3 = 0.06351 \cdot p^2 - 0.9416 \cdot p + 526.4 \qquad (33)$$

$$\nu^* = 0.003307 \cdot p^2 - 0.7862 \cdot p + 65.58 \qquad (34)$$

p represents percentage of PbO.

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* For Part I see *Jour. Amer. Ceram. Soc.*, **23** [11] 315–26 (1940).

[57] E. Zschimmer, "Physical Properties of Glasses as a Function of Chemical Composition," *Z. Electrochem.*, **11** [38] 629–38 (1905).

[58] Articles by C. J. Peddle, *Jour. Soc. Glass Tech.*, Vol. 4 (1920) and Vol. 5 (1921).

[59] C. J. Peddle, "Development of Various Types of Glass: II, Interaction of Silica, Lime, and Sodium Oxide," *ibid.*, **4**, 20–45 (1920); p. 30.

[60] A. N. Finn and H. G. Thomson, "Density and Index of Refraction of Glass vs. Its Composition," *Jour. Amer. Ceram. Soc.*, **8** [8] 505–13 (1925).

[61] W. E. S. Turner, "Physical Properties of Glasses; Relation to Chemical Composition and Mode of Preparation," *Jour. Chem. Soc.*, **129**, 2091–2116 (1926); *Ceram. Abs.*, **6** [3] 101 (1927).

[62] S. English and W. E. S. Turner, "Physical Properties of Boric Oxide Containing Glasses and Their Bearing on General Problems of Constitution of Glass," *Jour. Soc. Glass Tech.*, **7** [26] 155 (1923); *Ceram. Abs.*, **3** [2] 36 (1924).

[63] O. Knapp, "Constituents and Advance Calculation of Soda-Borosilicate Glasses," *Glastech. Ber.*, **8** [3] 154–58 (1930); *Ceram. Abs.*, **10** [6] 417 (1931).

[64] (*a*) T. Takamatsu, "Relation Between Optical Constants and Composition of Glass: I, Soda-Lime Glass," *Repts. Imp. Ind. Research Inst., Osaka, Japan*, **6** [9] 1–36 (1925); *Ceram. Abs.*, **5** [10] 306 (1926).

(*b*) T. Takamatsu, "Relation of Composition of Glass to Its Optical Constants, II," *Repts. Imp. Ind. Research Inst., Osaka, Japan*, **8** [5] 1 (1927).

(*c*) T. Takamatsu, "III," *ibid.*, **9** [12] 1 (1929); *Ceram. Abs.*, **8** [9] 646 (1929).

[65] H. Mees, "Calculation of Mean Refractive Index and the ν Value of Silicate-Flint Glasses," *Sprechsaal*, **58**, 293 (1925).

* ν = Abbé value (dispersive index) = $\dfrac{n-1}{n_F - n_C}$.

n = refractive index of glass with respect to sodium *D*-line.

n_F = refractive index of glass with respect to hydrogen F-line.

n_C = refractive index of glass with respect to hydrogen *C*-line.

These formulas held good in commercial practice, provided the ratio of alkali to lead oxide content conformed to Zschimmer's rule for lead glasses, that is,

$$R_2O = (76 - p) \times 0.27.$$

For two glasses whose PbO contents differed by not more than 8%, equation (35) was used to calculate the refractive index of an intermediate composition.

$$n_{\text{intermediate}} = (n_A - n_B) \cdot p_A + n_B \qquad (35)$$

wherein p_A = weight fraction of glass A in the intermediate,
n_A and n_B = respective indices of refraction of components A and B.

Karmaus[66] showed that the linear equation (36) was not sufficiently accurate for the calculation of refractive index from composition. His experimental values for the constants, n_X, are given in Table XIII.

$$n = p_A \cdot n_A + p_B \cdot n_B \qquad (36)$$

wherein p_A and p_B = weight percent of A($PbSiO_3$) and B(various silicates)
n_A and n_B = their partial refractive indices.

TABLE XIII

CONSTANTS FOR REFRACTIVE INDEX CALCULATION AFTER KARMAUS[66]

$PbSiO_3$	1.8908	$K_2O \cdot 6SiO_2$ 1.4851	$K_2O \cdot 8SiO_2$ 1.4785
$K_2O \cdot 4SiO_2 \cdot 2B_2O_3$	1.5008	$Na_2O \cdot 6SiO_2$ 1.4808	

From a consideration that the refractive index of glass is an additive property of the refractive powers of the glass-forming oxides, Knapp[67] expanded equation (36) to include additional components and derived the constants listed in Table XIV.

TABLE XIV

CONSTANTS FOR REFRACTIVE INDEX CALCULATION AFTER KNAPP[67]

SiO_2	0.01467	CaO	0.01776
Na_2O	0.01575	MgO	0.01655
K_2O	0.01551	Al_2O_3	0.01581

The average deviation of experimental from calculated values was 0.1 to 0.2% for fourteen alkali-lime-silica glasses.

Discarding his former hypotheses, Knapp[68] suggested that glass is a solution of free silica in high-order silicates or a mutual solution of both. From this consideration, he calculated refractive index from an expanded form of equation (36), wherein p_A, p_B, etc., now represent mole percentages of silicates, and n_A, n_B, etc., are the corresponding constants of refraction. From a similar equation for mean dispersion,

$$(n_C - n_F) = p_A \cdot n'_A + p_B \cdot n'_B + \ldots . \qquad (37)$$

the dispersive index or Abbé value, ν, could be evaluated from the relationship,

$$\nu = \frac{-n + 1}{n_C - n_F}.$$

Refraction constants, n_X, and mean dispersion factors, n'_X (Table XV), were evaluated from data after Peddle.

Peddle did not analyze his glasses, and the application of his data to the foregoing equations and factors produced mean deviations between experiment and calculation of 0.3% for refractive index and 1% for mean dispersion.

TABLE XV

FACTORS FOR CALCULATION OF REFRACTIVE INDEX AND MEAN DISPERSION AFTER KNAPP

	Refraction factors × 10^4				Mean dispersion factors × 10^7			
	Silicates				Silicates			
	Mono-	Di-	Tri-	Tetra-	Mono-	Di-	Tri-	Tetra-
Na-	1517	1500	1497	1490	1017	909	847	814
K-	1504	1497	1491	1495	937	904	857	830
Ca-	1612	1571	1533	1525	1105	955	848	820
Mg-	1569	1514	1477	1490	935	814	770	794
Zn-	1584	1561	1533	1535	1049	1003	926	894
Ba-	1589	1613	1580	1560	989	1094	884	994
Al-			1482				742	
SiO_2		1509				820		

Faick and Finn[69(a)] measured refractive indices to the fourth decimal for twenty-one soda-silica and twenty-one soda-lime-silica analyzed glasses. From their data, the authors[69(b)] first derived equation (38), correlating refractive index with composition.

$$n = n'_{SiO_2} + n'_{Na_2O} + n'_{CaO} \qquad (38)$$

wherein n = refractive index of glass
values of n' = complex functions of component percentages.

Later, from a consideration of refractive index-percentage silica curves, equation (39) was derived to apply to soda-silica glasses:

$$(n - 1) = (n'_A - 1) + (n'_B - 1) \qquad (39)$$

wherein
$$(n'_A - 1) = 0.005941 \cdot p_A - 0.000013543 \cdot p^2_A \qquad (40)$$

$$(n'_B - 1) = 0.005054 \cdot p_B. \qquad (41)$$

p_A and p_B = component percentages
n'_A and n'_B = their respective contributions to total index of refraction.

In this derivation, Faick and Finn assumed that soda had a linear effect on refractive index and any graphic curvatures should be attributed to silica. Their defini-

[66] H. Karmaus, "Studies on Some Chemical and Physical Properties of Lead Glasses as Functions of Their Composition," *Sprechsaal.*, **59**, 725, 747, 768 (1926); *Ceram. Abs.*, **6** [8] 336 (1927).

[67] O. Knapp, "Brilliancy and Composition of Glassware," *Glass Ind.*, **8** [4] 73–74 (1927); *Ceram. Abs.*, **6** [6] 215 (1927).

[68] (*a*) O. Knapp, "Constitution of Glass Based on Laws of Solution," *Jour. Soc. Glass Tech.*, **15** [58] 140–52 (1931); *Ceram. Abs.*, **10** [10] 683 (1931).
(*b*) O. Knapp, "Refraction of Glasses as a Constitutive Property," *Sprechsaal*, **63** [4] 61–64 (1930); *Ceram. Abs.*, **9** [6] 417 (1930).

[69] (*a*) C. A. Faick and A. N. Finn, "Index of Refraction of Some Soda-Lime-Silica Glasses as a Function of the Composition," *Jour. Amer. Ceram. Soc.*, **14** [7] 518–28 (1931).
(*b*) Anon., "Relations Between Chemical Composition, Index of Refraction, Density, and Thermal Expansion of Soda-Lime-Silica Glasses," *Ceram. Age*, **17** [3] 133 (1931).

tion of n' as a partial index should be avoided because the symbol actually represents an apparent index of refraction.

An extension of equation (39) produced equations (42) and (43) for the soda-lime-silica system.

$$(n-1) = (n'_A - 1) + (n'_B - 1) + (n'_C - 1) \quad (42)$$

wherein $$(n'_C - 1) = 0.00707 \cdot p_C. \quad (43)$$

The average error between measured and calculated values for forty-two soda-lime-silica glasses was 0.04%, the maximum deviation being 0.12%.

Later study indicated to Faick and Finn that refractive index-percentage SiO_2 plots for Na_2O-SiO_2 glasses could best be represented by three linear segments. The two intercepts occurring at 59.5 and 73.75% silica correspond closely to the compounds $2Na_2O \cdot 3SiO_2$ and $Na_2O \cdot 3SiO_2$. The straight-line hypothesis was then applied to the calculation of refractive index, and the general equation (44) for soda-silica and soda-lime-silica glasses became

$$(n-1) = a \cdot p_A + b \cdot p_B + c \cdot p_C \quad (44)$$

wherein p_A, p_B, and p_C = percentages of silica, soda, and lime, respectively
a, b, and c = empirical constants with values listed in Table XVI.

TABLE XVI

CONSTANTS FOR CALCULATION OF REFRACTIVE INDEX AFTER FAICK AND FINN

SiO_2 (%)	(a)	(b)	(c)
50? – 59.5	0.004836	0.005491	0.007521
59.5 – 73.75	0.004785	0.005568	0.007598
73.75–100	0.004584	0.006127	0.007977

Faick and Finn, applying the data of Turner for soda-lime-silica glasses to equations (42) and (44), found a mean error of less than 0.05% and a maximum error of 0.13%. A similar test using data after Peddle did not produce satisfactory agreement, a result possibly attributable to the fact that his glasses were not analyzed. Methods similar to those for soda-silica glasses were applied to eighty-six potash-silica glasses.[70] For a plot of refractive index against potash or silica content, the data could best be symbolized by two straight lines which intersected at 29% K_2O and were represented by equations (45) and (46).

$$n = 1.4676 + 0.00092 \cdot p \quad (45)$$

$$n = 1.4603 + 0.00118 \cdot p' \quad (46)$$

wherein n = refractive index for potash-silica glass
p and p' = percentages of K_2O greater and less than 29%, respectively.

Gilard and Dubrul,[71] in an extensive study parallel to that on density (see Part I, p. 319), formulated equation (47).

$$n \cdot \Sigma(p/100) = \Sigma(a \cdot p + b \cdot p^2) \quad (47)$$

wherein n = refractive index of glass
p = weight percentage of any oxide
a and b = appropriate constants.

The refraction coefficient for silica was assumed to be constant and from it corresponding values for other oxides were additively obtained (Table XVII). Gilard and Dubrul again used the adjective "partial" for "apparent" in describing the refractive index of any constituent.

They also indicate that errors of the following magnitude may be expected: (i) 0.1%, owing to refractive-index measurements to the third decimal only; (ii) 0.1%, caused by imperfect annealing of the test glasses; and (iii) 0.4%, owing to errors in analyses.

The total error of 0.6% represents a unit deviation of 0.009 for a glass of $n = 1.500$. The accuracy of microscopic measurements is of the order of 0.005 of a refractive index unit.

[70] "Index of Refraction of Some Potash-Silica Glasses," *Bur. Stand. Tech. News Bull.*, No. 224, pp. 123–24 (1935).

[71] Pierre Gilard and Léon Dubrul, "Calculation of Physical Properties of Glass: III, Index of Refraction," *Jour. Soc. Glass Tech.*, **21** [88] 476–88 (1937); *Ceram. Abs.*, **17** [9] 301 (1938).

TABLE XVII

REFRACTIVE INDEX CONSTANTS AFTER GILARD AND DUBRUL[71]

Oxides	$(a \cdot p + b \cdot p^2)$	Limits	Errors (%)
Li_2O	$0.01820 \cdot p$	0– 25	1.0
	$0.01980 \cdot p - 64 \times 10^{-6} \cdot p^2$	25– 45	1.0
Na_2O	$0.01618 \cdot p$	0– 20	
	$0.01646 \cdot p - 14 \times 10^{-6} \cdot p^2$	20– 55	0.2
K_2O	$0.01575 \cdot p$	0– 40	0.5 without PbO
	$0.01575 \cdot p$	0– 20	1.0 with PbO
	$0.01575 \cdot p + 130 \times 10^{-6} \cdot (p - 20)^2$	20– 60	1.0 with PbO
BeO	$0.01860 \cdot p - 80 \times 10^{-6} \cdot p^2$	0– 16	0.5
MgO	$0.01700 \cdot p$	0– 15	0.4
CaO	$0.01785 \cdot p$	0– 40	0.5
BaO	$0.01690 \cdot p$	0– 50	0.5
ZnO	$0.01675 \cdot p$	0– 18	0.1
PbO	$0.01760 \cdot p$	0– 40	1.0
	$0.01760 \cdot p + 140 \times 10^{-6} \cdot (p - 40)^2$	40– 80	1.0
B_2O_3	$0.01725 \cdot p - 50 \times 10^{-6} \cdot p^2$	0– 40	1.5
Al_2O_3	$0.01510 \cdot p$	0– 20	0.2
Fe_2O_3	$0.01875 \cdot p$	0– 15	
	$0.01875 \cdot p + 88 \times 10^{-6} \cdot (p - 15)^2$	15– 40	
Bi_2O_3	$0.01664 \cdot p + 46 \times 10^{-6} \cdot p^2$	0– 50	0.3
SiO_2	$0.01458 \cdot p$	0–100	
TiO_2	$0.0200 \cdot p$	0– 10	0.08

Knapp[72] used accurate composition and refractive index data for sixty common optical glasses to check the validity of the Gilard and Dubrul constants. A comparison of measured and calculated refractive indices indicated that agreement beyond the second decimal place could not be expected.

(B) Calculations Involving a Factor of Volume or Density

Tillotson[73] proposed the use of volume percentage rather than weight percentage in linear additive calculations of refractive index. From a mathematical derivation involving light velocity, refractive index, and time, he obtained,

$$n = q_1 \cdot n_1 + q_2 \cdot n_2 + q_3 \cdot n_3 + \ldots . \qquad (48)$$

wherein q_1, q_2, etc. = volume percentages.

Application of equation (48) to solutions and mixtures of liquids obtained linear refractive index-volume relations for approximately one hundred cases.

Tilley,[74] assuming additivity of specific refractivity, also indicated mathematically that refractive index is dependent on specific volume rather than on weight.

Factors of specific refractivity for various oxides and silicates in glass are available. Because specific refractivity involves both density and refractive index, a knowledge of the first two factors enables the latter to be calculated. This point will be discussed later under composition-refractivity relations (section IV).

Equations connecting specific volume and refractive index for some soda-silica, soda-lime-magnesia-silica, and potash-silica glasses were formulated at the Bureau of Standards.[75] The empirical nature of the expressions rendered them of little significance. A later publication[76] offered formula (49) by which refractive index in relation to other wave lengths might be calculated.

$$n_w = n_q + K(0.4540 - v) + K' \cdot p \qquad (49)$$

wherein n_w = refractive index for a certain wave length, w
n_q = refractive index of fused quartz for same wave length
v = specific volume of glass
p = lime content of glass
K and K' = constants whose values are determined by $K = 0.8164 + 2{,}780{,}000 \cdot D$ and $K' = 0.00128 - 4274 \cdot D$

wherein D = square of vibration number for wave length, w.

The calculated values were within the errors of measurement and held for the region of wave lengths from 7682 Å to 4047 Å.

(C) Graphic Representation

Wright[77] prepared several graphs from which could be read the percentages of PbO, SiO_2, and alkali oxides required to produce a definite index of refraction.

Morey and Merwin[78] and Faick and Finn[79] formulated iso-refractive index curves for the system soda–lime–silica, so that the refractive index could be predicted from the composition.

From Tillotson's conclusions that volume rather than weight percentage indicates linear additivity, it is suggested that the former should be adopted for graphic representation. It may also be preferable to use weight rather than mole percentage because refractive index is related more closely to weight than to the number of molecules.

IV. Relation Between Composition and Specific or Molecular Refractivity

It is generally true that the refractive index of glass varies directly with density,[80] although the relation is not a simple one. Tilley[73] observed, from refractive index-density plots, that natural-occurring glasses were grouped into distinct fields. Investigators have long sought a simple refractive index-density relation that would be independent of temperature, pressure, and concentration. Specific refractivity* is that quantity which represents this relation. Atomic and molecular refractivities are the products of atomic or molecular weights and specific refractivities. The expressions for specific refractivity commonly encountered in glass technology follow in equations (50) through (54).

(*a*) Newton-Laplace formula,[81]

$$k = \frac{n^2 - 1}{d} \qquad (50)$$

(*b*) Gladstone-Dale formula,[82]

$$k = \frac{n - 1}{d} \qquad (51)$$

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[72] O. Knapp, "Calculation of Refractivity of Glasses," *Jour. Soc. Glass Tech.*, **24** [1] 37–40 (1940); *Ceram. Abs.*, **19** [8] 100 (1940).

[73] E. W. Tillotson, Jr., "Relation Between Physical Properties and Chemical Composition of Glass: VIII, Molecular Compounds," *Jour. Amer. Ceram. Soc.*, **1** [2] 76–93 (1918).

[74] C. E. Tilley, "Density, Refractivity, and Composition Relations of Some Natural Glasses," *Mineralog. Mag.*, **19** [96] 275–94 (1922).

[75] "Research Work in Glass Industry Reported by Bureau of Standards," *Ceram. Ind.*, **18** [2] 92–97 (1932).

[76] "Density and Refractivity of Some Soda-Lime Glasses" (note from Bureau of Standards), *Jour. Franklin Inst.*, **214** [6] 751–52 (1932).

[77] (*a*) F. E. Wright, "Certain Relations Between Chemical Composition and Refractivity in Optical Glass," *Jour. Amer. Ceram. Soc.*, **3** [10] 783–832 (1920).

(*b*) F. E. Wright, Manufacture of Optical Glass and of Optical Systems, Ordnance Department Document No. 2307. U. S. Government Printing Office, Washington, D. C., 1921; *Ceram. Abs.*, **1** [3] 93 (1922).

[78] G. W. Morey and H. E. Merwin, "Relation Between Composition and Density and Optical Properties of Glass: I, Soda-Lime-Silica Glasses," *Jour. Optical Soc. Amer.*, **22**, 632–62 (1932); p. 656; *Ceram. Abs.*, **12** [6] 218 (1933).

[79] See footnotes 69 and 75.

[80] H. Schulz, Das Glas, p. 115. J. Kosel and F. Pustet Komm-Ges., München, Germany, 1923; *Ceram. Abs.*, **3** [9] 262 (1924).

* Otherwise called specific refraction, specific refractive power, or specific refractive energy.

[81] P. Simon Marquis de Laplace, Mécanique Céleste, (translated by N. Bowditch), Vol. 4, p. 454. Hilliard, Gray, Little, and Wilkins, Boston, Mass., 1829–1839.

[82] (*a*) J. H. Gladstone and T. P. Dale, "Researches on Refraction, Dispersion, and Sensitiveness of Liquids," *Phil. Trans. Roy. Soc.* (London), **153** [I] 317–43 (1863).

(*b*) T. Preston, Theory of Light, 4th ed., p. 138. Macmillan & Co., London, 1912.

(*c*) Lorentz-Lorenz formula,[83]

$$k = \frac{n^2 - 1}{n^2 + 2} \cdot \frac{1}{d} \qquad (52)$$

(*d*) Eykman formula,[84]

$$k = \frac{n^2 - 1}{n + 0.4} \cdot \frac{1}{d} \qquad (53)$$

(*e*) Lichtenecker formula,[85]

$$k = \frac{\log n}{d} \qquad (54)$$

wherein k = specific refractivity
n = refractive index
d = density.

Equation (50) after Newton-Laplace shows poor agreement with experimental data and will not be discussed.

Quoting Gladstone and Dale,[82(c)] "Every liquid has a specific refractive energy, composed of the specific refractive energies of its component elements modified by the manner of combination, which is unaffected by changes of temperature and accompanies it when mixed with other liquids. The product of this specific refractive energy and the density is, when added to unity, the refractive index." This statement in mathematical form yields equation (55).

$$k = (1/100) \cdot (p_A \cdot k_A + p_B \cdot k_B + \ldots) \qquad (55)$$

wherein k = specific refraction of any substance, k_A, k_B p_A, p_B = specific refractions and weight percentages of components A and B.

In addition to changes in volume that can occur on mixing two liquids, there may be similar changes in specific refractive powers which may or may not be proportional to the volume variations. Schütt[86] and Pulfrich[87] pointed out that the specific refraction of mixtures of two liquids is not necessarily a linear function of that of the components, and to express these relations, they derived complex formulas which showed excellent agreement with experimental results.

Assuming linear additivity of specific volume, Tillotson[88] showed that the formulas of Schütt and Pulfrich could be reduced to

$$k = \Sigma q_X \cdot k_X.$$

He expected that the specific refractive power of a mixture of silicates would be a nearly linear function of the components expressed in percentages by volume.

In organic chemistry, the addition of atomic refractivities to obtain molecular refractivities has been used for determinations of compound structures.[89] A clear concept of refractive energy, however, is lacking. Herz[90] found that the quotient of the parachor and the molecular refraction as determined by the Lorentz-Lorenz formula was a constant, independent of the liquids examined. According to Clausius-Mosotti-Exner,[91] molecular refraction calculated by means of the Lorentz-Lorenz formula represents the true volume of one mole of a substance. This opinion was substantiated by Herz,[92] who calculated for a large number of compounds the product of molecular refraction and the number of moles per milliliter at the boiling point. Except for some low boiling substances, an approximately constant value of (1.2 to 2) $\times 10^{23}$ was obtained for fifty compounds.

(A) Additive Calculations—Oxide and Compound Factors

Larsen[93] tested the Gladstone-Dale and Lorentz-Lorenz formulas with the data from six albite ($Na_2O \cdot Al_2O_3 \cdot 6SiO_2$)–anorthite($CaO \cdot Al_2O_3 \cdot 2SiO_2$) and twelve $CaSiO_3$–$MgSiO_3$ glasses. Of the properties of refractive index, specific volume, and specific refractivity, the last alone showed additivity. Mean deviations between measured and calculated refractivities for the two respective formulas were 0.06 and 0.07% in the case of feldspar glasses and 0.12 and 0.14% for the calcium-magnesium-silicate glasses.

Larsen and Berman[94] applied the Gladstone-Dale formula in the calculation of specific refractive energies for the chief constituents of crystalline minerals by using the mean refractive index and the density of the crystals. They obtained values for sixty metallic and nonmetallic oxides and eight nonmetallic elements. For a number of minerals selected at random, computed and experimental data agreed within 5%.

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[83] (*a*) H. A. Lorentz, "Relation Between Propagation of Light and Density of Matter," *Wied. Ann. Phys. Chem.* [Series 9], **9** [4] 641–65 (1880).
(*b*) L. Lorenz, "Refraction Constants," *ibid.*, [Series [9], **11** [9] 70–103 (1880).
(*c*) F. G. Getman and F. Daniel, Outline of Theoretical Chemistry, 6th ed., p. 632. John Wiley & Sons, Inc., New York, N. Y., 1938.

[84] J. F. Eykman, "Refractometric Observations," *Rec. trav. chim.*, **14**, 185–202 (1895).

[85] (*a*) K. Lichtenecker, "Calculation of Optical Refractive Indices by Logarithmic Rule of Averages," *Physik. Z.*, **26**, 297–305 (1925).
(*b*) K. Lichtenecker, "Theory of Mixtures and the Logarithmic Mixing Rule," *ibid.*, **27**, 833–37 (1926).

[86] F. Schütt, "Determination of Molecular Refraction of Solid Compounds in Solution," *Z. physik. Chem.*, **5** [4] 349–73; **9** [3] 349–77 (1892).

[87] C. Pulfrich, "Refractive Power of Mixtures of Two Liquids," *ibid.*, **4** [5] 561–69 (1889).

[88] E. W. Tillotson, Jr., "Relation of Refractive Index of Soda-Barium and Soda-Lime Glasses to Their Chemical Composition," *Jour. Ind. Eng. Chem.*, **4** [11] 882–85 (1912).

[89] (*a*) S. Smiles, Relation Between Chemical Constitution and Some Physical Properties, chapters viii and ix. Longmans, Green, & Co., New York, N. Y., 1910.
(*b*) J. B. Cohen, Organic Chemistry for Advanced Students, 5th ed., Vol. II, pp. 19–45. Longmans, Green, & Co., London, 1928.
(*c*) H. Gilman, Organic Chemistry, an Advanced Treatise, Vol. II, pp. 1737–42. John Wiley & Sons, Inc., New York, N. Y., 1938.

[90] W. Herz, "Molecular Refraction and Parachor," *Z. anorg. allgem. Chem.*, **159** [4] 316–18 (1927).

[91] F. Exner, "New Method for Determination of Molecular Dimensions," *Wiener Monatshefte für Chemie*, **6**, 249 (1885).

[92] W. Herz, "Molecular Refraction and Molecular Number," *Z. anorg. allgem. Chem.*, **179** [1–3] 211–14 (1929).

[93] E. S. Larsen, "Relation Between Refractive Index and Density of Some Crystallized Silicates and Their Glasses," *Amer. Jour. Sci.* [4th Series], **178** [28] 263–74 (1909).

[94] E. S. Larsen and H. Berman, "Microscopic Determination of the Nonopaque Minerals," *U. S. Geol. Surv. Bull.*, No. 848, 2d ed., 254 pp. (1934); p. 30; *Ceram. Abs.*, **13** [8] 221 (1934); for review see *Ceram. Abs.*, **13** [7] 196 (1934).

Tillotson,[95] with the aid of density factors and measured refractive indices, calculated specific refractivities, $k_{obs.}$, for twelve soda lime-silica glasses. The so-called $k_{calc.}$ was obtained from smooth curves, representing $k_{obs.}$ against percentage of $CaSiO_3$. From calculated values of k, computed refractive indices were derived. Average differences in refractive index for experiment and calculation were 0.06 and 0.09% respectively, for the Gladstone-Dale and Lorentz-Lorenz formulas. When using the expression of Gladstone-Dale, specific refractivity-weight percentage composition curves indicated marked breaks at the composition point corresponding to $2Na_2O \cdot 3CaO \cdot 5SiO_2$. The following constants of specific refractivity were derived from these curves to fit equation (55):

SiO_2	0.1220	CaO	0.1210	Na_2O	0.1302

Tillotson[88] carried out similar studies on four series of ten glasses each for the system soda–barium oxide–silica. In the range $Na_2O \cdot 3SiO_2$–$BaO \cdot 2SiO_2$, a break corresponding to the compound $Na_2O \cdot BaO \cdot 5SiO_2$ resulted from a plot of specific refraction against volume percentage of $BaO \cdot 2SiO_2$. Mean deviations between calculated and observed refractive indices for the four series were, respectively, 0.0019, 0.0026, 0.0012, and 0.0010 unit. As different expressions of composition were used in the two studies, it is not surprising that the following constants of specific refraction do not agree with the values first obtained:

SiO_2	0.1995	CaO	0.2410
BaO	0.0500	Na_2O	0.2360

From data after Peddle, Stott[96] calculated specific refractions for soda-silica glasses and found, on plotting the values against weight percentage of Na_2O, a marked break corresponding to the compound $Na_2O \cdot 2SiO_2$. Similar specific volume-composition curves were smooth. Because specific volume is involved in the specific-refraction formula, Winks and Turner[97] suggested that refractive index and not density is a criterion of constitution. Faick and Finn,[76] however, observed that the breaks in refractive index-composition curves for soda-silica glasses corresponded to the compounds, $2Na_2O \cdot 3SiO_2$ and $Na_2O \cdot 3SiO_2$.

Tilley[75] was able to separate tektite and volcanic glasses by plotting specific refractivities (computed according to the Gladstone-Dale formula) against densities. The specific refractivities of five analyzed glasses showed good agreement with values calculated from the specific refractivities of the components.

Bannister,[98] in an extension of Tilley's method to classify artificial glasses, plotted refractive index against density and obtained three straight lines of different slopes for Peddle's glasses of the general formula, $100SiO_2 \cdot 20Na_2O \cdot xRO$, where RO may be CaO, BaO, or PbO. These lines, which could be expressed by equation (56), tended to converge toward a single point theoretically corresponding to vitreous silica. A plot of a against b or a graph of the ratio of apparent refractive index to apparent density of Na_2O and RO against n or d allowed satisfactory grouping of certain artificial gem stones and various glasses after Morey and Wright. Thallium and lead glasses were separated by plotting refractive index against dispersion, a method commonly used to classify optical glasses.[99]

$$n = a \cdot d + b \qquad (56)$$

wherein a and b = constants.

In a study of specific refraction-weight composition curves for the series, $20\ Na_2O \cdot (80 - x)\ SiO_2 \cdot xB_2O_3$, English and Turner[100] observed breaks corresponding to the compound, $3SiO_2 \cdot Na_2O \cdot B_2O_3$. For the series, $10Na_2O \cdot (90 - x)SiO_2 \cdot xB_2O_3$, curve breaks represented the compound, $8SiO_2 \cdot Na_2O \cdot B_2O_3$. The existence of these 1:1 B_2O_3–Na_2O ratios supports the explanation given by Warren[101] for the abnormal behavior of soda-boric oxide-silica glasses because the slopes of the lines change at the point where all boron atoms are in tetrahedral coordination.

Turner and Winks,[102] employing the Lorentz-Lorenz formula, calculated the specific refractivities for ten glasses of the formula, $(75.8 - x)SiO_2, xB_2O_3, 8.56CaO, 6.86Na_2O, 7.9K_2O$, wherein x varied from 0 to 40%. Specific refraction-percentage B_2O_3 plots indicated that linear graphs of two segments should be drawn, although the plotted points showed poor coincidence. A change of curve direction was evident at 7% B_2O_3.

Eckert,[103] in a test of the Gladstone-Dale and Lorentz-Lorenz formulas, found no simple linear relation between specific refraction and volume percentage of PbO for lead glasses. The same formulas were employed by Karmaus[66] to calculate specific refractivities of the system $x(PbO \cdot SiO_2) \cdot (10 - x)(K_2O \cdot 6SiO_2)$. Both methods of calculation obtained refraction-volume percentage $PbO \cdot SiO_2$ curves of two segments.

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[95] E. W. Tillotson, "Relation of Refractive Index of Soda-Lime Glasses to Their Chemical Composition," *Jour. Ind. Eng. Chem.*, **4** [4] 246–49 (1912).

[96] V. H. Stott, discussion of paper by C. J. Peddle, "Development of Optical Glass During the War," *Jour. Soc. Glass Tech.*, **4**, 232 (1920).

[97] F. Winks and W. E. S. Turner, "Density of Sodium Metasilicate-Silica Glasses," *ibid.*, **15** [58] 185–90 (1931); *Ceram. Abs.*, **10** [9] 630 (1931).

[98] F. A. Bannister, "Relation Between Density and Refractive Index of Silicate Glasses, with Application to Determination of Imitation Gem-Stones," *Mineralog. Mag.*, **22** [126] 136–54 (1929); *Ceram. Abs.*, **9** [5] 342 (1930).

[99] O. Knapp, "Optical Glasses," *Glass Ind.*, **19** [12] 465–68 (1938); p. 467; *Ceram. Abs.*, **18** [5] 125 (1939).

[100] S. English and W. E. S. Turner, "Density of Soda-Lime-Magnesia Glasses and Calculation of Density," *Jour. Soc. Glass Tech.*, **6**, 228–31 (1922); *Ceram. Abs.*, **2** [10] 219 (1923); see also footnote reference 62.

[101] (*a*) J. Biscoe and B. E. Warren, "X-Ray Diffraction Study of Soda-Boric Oxide Glass," *Jour. Amer. Ceram. Soc.*, **21** [8] 287–93 (1938).

(*b*) J. Biscoe, C. S. Robinson, Jr., and B. E. Warren, "X-Ray Study of Boric Oxide-Silica Glass," *ibid.*, **22** [6] 180–84 (1939).

[102] W. E. S. Turner and F. Winks, "Influence of Boric Oxide on Properties of Chemical and Heat-Resisting Glasses: I, Preparation and Physical Properties of the Glasses," *Jour. Soc. Glass Tech.*, **9** [36] 389–405 (1925); *Ceram. Abs.*, **5** [5] 145 (1926).

[103] F. Eckert, "Physical Properties of Glass," *Jahrb. Radioakt. Elektronik*, **20**, 93–275 (1923); *Ceram. Abs.*, **3** [8] 245 (1924).

For the calculation of refractive index of lead glasses, Karmaus studied four additive expressions, which singly represented considerations of volume and weight percentages and refraction computed by the formulas of Lichtenecker and Lorentz-Lorenz. He found that the best expression of linear additivity in equations involved the use of volume rather than of weight percentage, an observation apparently coinciding with the theory of Tillotson (see Part I, p. 346). The best value of refractive index for $PbSiO_3$ (1.8908) resulted from equations (57) and (58).

$$k = q_A \cdot k_A + q_B \cdot k_B \tag{57}$$

$$\text{Log } n = q_A \cdot \log n_A + q_B \cdot \log n_B \tag{58}$$

wherein q = volume percentage
k = specific refraction determined from the Lichtenecker formula
A and B = components.

From these data and independently by the method of Mees,[65] Karmaus calculated refractive indices for the glass series $x(PbSiO_3)\cdot(10 - x)(K_2O\cdot 6SiO_2)$, the average deviation from experiment being 0.04% in both cases. Lichtenecker's logarithmic rule of mixtures is thus of value for an estimation of the optical properties of lead glasses.

Data after Peddle were used by Knapp[68(b)] to calculate molecular refractions from equation (59).

$$K = (1/100)\cdot(P_A \cdot K_A + P_B \cdot K_B) \tag{59}$$

wherein K = molecular refraction expressed by the Lorentz-Lorenz formula
P_A and P_B = mole percentages of SiO_2 and Na_2SiO_{2n+1}, respectively
K_A and K_B = molecular refractions for components (values given in Table XVIII).

TABLE XVIII

MOLECULAR REFRACTION FACTORS AFTER KNAPP

Compounds	Differences	Mol. refraction	Mol. refraction difference /SiO_2
SiO_2		7.471	
Na-monosilicate	SiO_2	14.51	7.04
Na-disilicate	SiO_2	21.73	7.22
Na-trisilicate	SiO_2	29.33	7.60
Na-pentasilicate	$2SiO_2$	44.27	7.47

The mean difference in the value of molecular refraction for each successive increment of SiO_2 is 7.33. Calculated molecular refractions deviated about 1.2% from values computed from experimental data. Knapp derived the specific refractivity of CaO (0.114), and deviations of the order of 1% were obtained in a test of soda-lime-silica glasses in which $Na_2Si_nO_{2n+1}$, $CaSi_nO_{2n+1}$, and SiO_2 were considered to be constituents.

From Table XVIII, Knapp calculated the following values for the atomic refractions of Na, Si, and O:

Na 2.6 Si 3.6 O 1.9

Morey and Merwin[73] evaluated for soda-lime-silica glasses, the following specific-refractivity expressions, based on the refraction equations (51), (52), (53), and (54). They assigned each oxide an arbitrary constant and assumed that specific refractivity is a linear function of composition. Except for the Lorentz-Lorenz formula, the constant used for silica was that calculated from the values of refractive index and density of vitreous silica. Factors for CaO and Na_2O were so chosen as to produce an even distribution of positive and negative deviations between experiment and computation.

(*a*) From the Gladstone-Dale formula,

$$100k = 0.1947\cdot p_N + 0.2268\cdot p_C + 0.2081\cdot p_S. \tag{60}$$

(*b*) From the Lorentz-Lorenz formula,

$$100k = 0.1115\cdot p_N + 0.1221\cdot p_C + 0.1237\cdot p_S. \tag{61}$$

(*c*) From the Eykman formula,

$$100k = 0.2530\cdot p_N + 0.2807\cdot p_C + 0.2753\cdot p_S. \tag{62}$$

(*d*) From the Lichtenecker formula,

$$100k = 0.6231\cdot p_N + 0.7508\cdot p_C + 0.7440\cdot p_S. \tag{63}$$

wherein k = specific refractivity of glass.
p_N, p_C, and p_S = weight percentages of Na_2O, CaO, and SiO_2, respectively.

Percentage deviations from additivity for equations (60) through (63), plotted on a triaxial diagram, showed similar patterns of departure. The best results were obtained from the Gladstone-Dale formula whose accuracy was $\pm 0.1\%$. The maximum volume contraction resulting from mixing of compositions was 4%.

Finn and his co-workers (see Part I, p. 319 and Part II, p. 345) plotted specific volume and refractive index of soda-silica glasses against weight percentage of SiO_2. Curves were segmented into three straight lines, each of which could be expressed by equations (64) and (65).

$$v = a\cdot p + b\cdot p' \tag{64}$$

and

$$n - 1 = a'\cdot p + b'\cdot p' \tag{65}$$

wherein v and n = specific volume and refractive index
p and p' = weight percentage of SiO_2 and Na_2O, respectively
a, b, a', and b' = corresponding constants (values given in Table VI (Part I, p. 320) and in Table XVI).

Because $p + p' = 100$, equations (64) and (65) may be written as follows:

$$v = 100\cdot b + (a - b)\cdot p. \tag{66}$$

$$n - 1 = 100\cdot b' + (a' - b')\cdot p. \tag{67}$$

Inasmuch as $k = (n - 1)/d = (n - 1)\cdot v$, wherein k is specific refractivity based on the Gladstone-Dale formula, the reviewers multiplied equations (66) and (67) to obtain equation (68) for calculating specific refractivities of soda-silica glasses.

$$k = 10^4 \times bb' + 100\,(b(a' - b') + b'(a - b))\cdot p + (a - b)(a' - b')\cdot p^2. \tag{68}$$

Suitable constants were chosen and equations (69), (70), and (71) were formulated to apply to the composition ranges of the segments of Finn's curves.

(*a*) 50 to 59% SiO_2,

$$k = 10^{-4}(19.14 + 0.02077\cdot p - 0.00005201\cdot p^2). \tag{69}$$

(*b*) 63 to 74% SiO_2,

$$k = 10^{-3}(18.52 + 0.03211 \cdot p - 0.00008178 \cdot p^2). \quad (70)$$

(*c*) 75 to 100% SiO_2,

$$k = 10^{-3}(17.34 + 0.06114 \cdot p - 0.0002639 \cdot p^2). \quad (71)$$

Because the results of Finn probably represent the most accurate data extant for soda-silica glasses, equations (69), (70), and (71) may be considered to be valuable experimental expressions for specific refractivity. The presence of a squared term indicates that specific refractivity is not a linear function of composition, an observation not represented in equation (60) of Morey and Merwin. The omission of such terms results in large errors. Where rapidity of calculation is desired, the additive equation (72) may be used,

$$100k = 0.1929 \cdot p_N + 0.2081 \cdot p_S. \quad (72)$$

Equations (69), (70), and (71) were assumed to be fundamentally correct, and deviations therefrom were of the order of 0.24 and 0.07%, respectively, for equation (60) after Morey and Merwin and equations (72) after the reviewers.

Jenckel[104] prepared ten glasses in the system B_2O_3–$Na_2B_4O_7$. The reviewers obtained a continuous concave curve from a plot of specific refractivity against weight percentage of $Na_2B_4O_7$, thus indicating apparent invalidity of linear additivity for Jenckel's data.

Biltz, Weibke, and Schrader-Traeger[105] found a linear addition relation for 400 glasses whose molecular refractions were calculated from the formula,

$$K = (1/100)(P_A \cdot K_A + P_B \cdot K_B + \ldots) \quad (73)$$

wherein K = molecular refraction expressed by the Gladstone Dale equation
P_A and P_B = mole percentages
K_A and K_B = corresponding molecular refraction factors for components A and B (Table XIX).

Molecular refractions for SiO_2 and other component oxides (Table XX) were derived from the solution of simultaneous equations involving alkali-silicate glasses of appropriate molecular compositions.

The deviations of calculated from measured molecular refractions for the glasses tested ranged from 0.07 to 0.35%.

A knowledge of molecular refractions enabled Biltz and his co-workers to calculate refractive indices from equation (74).

$$n = (K/V)_{calc.} + 1. \quad (74)$$

The accuracy of this expression was stated to be 0.3%. Actual computed indices for soda-silica and soda-lime-silica glasses showed deviations of 0.09 and 0.14%, respectively, when calculated values of V and K were used and of 0.04 and 0.04% when calculated values of K and measured values of V were used. Good agreement was also obtained for soda-boric oxide glasses, but the errors for the soda-lead oxide-silica and potash-lead oxide-silica glasses of Peddle ranged from 0.3 to 2%.

Young and Finn[106] have examined seven specific refraction ratios by a comprehensive treatment of available data. Their results indicated that of these ratios only the Gladstone-Dale value possesses the following desirable features: "(i) it varies linearly with chemical composition and with the square of the wave number of the incident light; (ii) for a given composition and wave length of light, it is substantially the same for both the vitreous and crystalline states; and (iii) for the vitreous state, it is independent of thermal history." Young and Finn indicate that their calculated Gladstone-Dale factors for forty-seven oxides are related to the periodicity of the elements.

(B) Calculation from Ionic Refractivities

The use of ionic refraction values is logical from a consideration of the modern concept of glass constitution. Randall and Gee[107] calculated the molecular refractivity of glass from ionic refractivities by equation (75), based on the Lorentz-Lorenz formula.

$$K = \frac{(n^2 - 1) \cdot M}{(n^2 + 2) \cdot d} = \Sigma K_i \quad (75)$$

wherein ΣK_i = summation of ionic refractivities
K, n, d, and M = molecular refraction, refractive index, density and mean molecular weight of glass, respectively.

They adopted certain values after Wasastjerna,[108] who in turn had applied the rule of Cuthbertson (relating atomic refractivities and atomic weights of gaseous elements) to calculate ionic refractivities in aqueous solution. These values and others, so chosen as to fit the equation best, are listed in Table XXI.

TABLE XIX

MOLECULAR REFRACTION FACTORS AFTER BILTZ, WEIBKE, AND SCHRADER-TRAEGER*

SiO_2 (glass)	12.502	$K_2Si_4O_9$	13.796
$Na_2Si_2O_5$	12.329	$K_2Si_2O_5$	14.665
Na_2SiO_3	12.264	$CaSiO_3$	12.56
Li_2SiO_3	9.64	$PbSiO_3$	21.860

* These values actually represent "equivalent" refractions.

TABLE XX

MOLECULAR REFRACTIONS OF GLASS-FORMING OXIDES AFTER BILTZ, WEIBKE, AND SCHRADER-TRAEGER

SiO_2	12.48	PbO	31.24
Na_2O	12.04	Al_2O_3	21.24
K_2O	19.05	Li_2O	6.8
CaO	12.64	B_2O_3	17.467

[104] Ernst Jenckel, "Constitution of Glasses," *Z. Elektrochem.*, **41** [7] 211–15 (1935); p. 214; *Ceram. Abs.*, **15** [8] 232 (1936).

[105] W. Biltz, F. Weibke, and L. Schrader-Traeger, "Molecular Refraction and Volumes of Glasses," *Z. anorg. allgem. Chem.*, **234**, 253 (1937); condensed article in *Glastech. Ber.*, **16** [4] 131–33 (1938); *Ceram. Abs.*, **17** [10] 326 (1938).

[106] J. C. Young and A. N. Finn, "Specific Refraction of Glasses and Glass-Forming Oxides," abstracted in *Glass Ind.*, **21**, 97 (1940).

[107] J. T. Randall and N. Gee, "Refractive Index, Atomic Size, and Packing in Glasses," *Jour. Soc. Glass Tech.*, **15** [57] 41–53 (1931); *Ceram. Abs.* **10** [8] 552 (1931).

[108] A. Wasastjerna, *Soc. Sci. Fennica, Commentationes Phys.-Math.*, **1** [37] 1 (1923).

TABLE XXI

IONIC REFRACTIVITIES (K_i)

O^{--}	3.9	Ca^{++}	2.0
Si^{4+}	0.25	Pb^{++}	11.65
Na^{+}	0.7	B^{3+}	0.05
K^{+}	2.85		

Refractive indices of twenty-eight glasses from nine systems were calculated, both from measured density values and from molecular refractions as determined by equation (75). Maximum divergency approximated 1.4%. A correlation of this study and their computations of volume from ionic radii enabled Randall and Gee to derive a new expression (equation (76)) for the direct calculation of refractive index.

$$\frac{(n^2 - 1)}{(n^2 + 2)} = \frac{\Sigma K_i}{1.244 \cdot \Sigma V_i} \qquad (76)$$

wherein ΣK_i and ΣV_i = summations of ionic refractivities and ionic volumes, respectively.

Tests of equation (76) indicated an accuracy of 1.4%.

Biltz, Weibke, and Schrader-Traeger[105] calculated the "ionic"* refractions of "frame" and "anion" oxygen in glass (Table XXII) in a manner similar to their computation of ionic volumes for these same oxygen types.

TABLE XXII

IONIC REFRACTION OF "FRAME" OXYGEN (BASED ON LORENTZ-LORENZ FORMULA) AFTER BILTZ, WEIBKE, AND SCHRADER-TRAEGER

Glass	Mol. refraction	Δ-mol. refraction of silicates	Ionic refraction of frame oxygen
SiO_2	7.447	7.447	3.72
Na_2SiO_3	14.355	7.368	3.68
$Na_2Si_2O_5$	21.723		
$K_2Si_2O_5$	25.818	2×(7.410)	3.71
$K_2Si_4O_9$	40.637		

Ionic refractivity of Si^{4+} is considered to be zero.

IONIC REFRACTION OF "ANION" OXYGEN

Glass	Ionic refraction	Glass	Ionic refraction
$Na_2O_2(SiO)$	4.84	$K_2O_2(Si_2O_3)$	5.07
$Na_2O_2(Si_2O_3)$	4.81	$K_2O_2(Si_4O_7)$	5.03

Table XXII indicates that the ionic refraction of "frame" oxygen is constant for different types of glass, whereas that of "anion" oxygen varies inversely as the ionic density of the binding ions (decrease in charge and increase in diameter of these ions).

Kordes[109] investigated glasses in the binary systems, SiO_2–Na_2O, SiO_2–PbO, B_2O_3–PbO, B_2O_3–ZnO, B_2O_3–CdO, B_2O_3–As_2O_3, B_2O_3–Sb_2O_3, B_2O_3–Na_2O, and P_2O_5–PbO. Except for the series P_2O_5–PbO and B_2O_3–Na_2O (up to 30 mole percentage of Na_2O), graphs relating molecular refraction, based on the Lorentz-Lorenz formula, and composition in mole percentage, were not linear for wide ranges of composition. Kordes indicates that the expression of Biltz[110] (equation (73)) must therefore have a limited scope. Nonlinearity was also observed in the soda–lime–silica system for glasses containing large amounts of silica or basic constituents. Because refraction constants in simple addition rules were found to be concentration-dependent, Kordes suggested that observed divergences from linearity were caused by oxygen ions whose volumes and ionic refractions were dependent on their mode of combination with other ions. He proposed the existence of three oxygen-ion types, the first two of which have already been mentioned.

(1) "Structure" or "frame" oxygens, linked on two sides to Si or B or P ions, were denoted by O^{Si}, O^{B}, and O^{P}, respectively. The ionic refraction of this type of oxygen was found to be independent of the concentration of SiO_2, B_2O_3, and P_2O_5 in glass.

(2) "Anion" oxygens, connected to Si or B or P ions on one side and such metallic cations as Na^+ or Ca^{++} on another side, were represented by O^{Si-M}, O^{B-M}, and O^{P-M}.

(3) Oxygen ions joined solely to metallic cations were denoted by O^{M}.

For types (2) and (3), ionic refractions were found to vary linearly with component concentrations.

As molecular refractions of glassy SiO_2, B_2O_3, and P_2O_5 were known and as ionic refractions of Si^{4+}, B^{3+}, and P^{5+} had been determined by Fajans and Joos,[111] Kordes was able to calculate ionic refraction values for O^{Si}, O^{B}, and O^{P} (Table XXIII).

TABLE XXIII

MOLECULAR AND IONIC REFRACTIONS

B_2O_3	10.45	B^{3+}	0.05	O^{B}	3.45
SiO_2	7.447	Si^{4+}	0.1	O^{Si}	3.673
P_2O_5	18.65	P^{5+}	0.07	O^{P}	3.70
PbO	15.50	Pb^{++}	9.13*	O^{Pb}	7.37
		Na^{+}	0.475*		

* L. Pauling, "Theoretical Prediction of Physical Properties of Many Electron Atoms and Ions," *Proc. Roy. Soc.* (London), **A114**, 181–211 (1927); p. 198.

An evaluation of ionic refractivities of the concentration-dependent factors, O^{Si-M} and O^{M}, was made by the empirical equations (77) and (78).

$$K_{O^{Si-M}} = P_1 \cdot K_1 + P_2 \cdot K_2 \qquad (77)$$

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* The word "ionic" is introduced by the reviewers.

[109] (*a*) E. Kordes, "Relation Between Fine Structure and Physical Properties of Glasses," *Glastech. Ber.*, **17** [3] 65–76 (1939); *Ceram. Abs.*, **18** [8] 207 (1939).
(*b*) E. Kordes, "Reply to Criticism," *Glastech. Ber.*, **17** [6] 187 (1939); *Ceram. Abs.*, **18** [11] 294 (1939).
(*c*) E. Kordes, "Physicochemical Investigation of Fine Structure of Glasses," *Z. anorg. allgem. Chem.*, **241** [1] 1–38 (1939).
(*d*) E. Kordes, "Reply to Biltz Note," *ibid.*, **241** [4] 418–20 (1939).

[110] (*a*) W. Biltz, "Relation Between Fine Structure and Physical Properties of Glasses, Criticism," *Glastech. Ber.*, **17** [6] 183 (1939); *Ceram. Abs.*, **18** [11] 294 (1939).
(*b*) W. Biltz and F. Weibke, "Note to Treatise on Molecular Refraction of Glasses," *Z. anorg. allgem. Chem.*, **241** [1] 39–41 (1939).
(*c*) W. Biltz and F. Weibke, "Concluding Note on Molecular Refraction of Glasses," *ibid.*, **241** [4] 421–23 (1939).

[111] K. Fajans and G. Joos, "Molrefraktionen von Ionen, etc.," *Z. Physik*, **23**, 1–47 (1924); p. 20.

$$K_{OM} = P_1 \cdot K_1 + P_2 \cdot K_O \quad (78)$$

wherein K_O = ionic refraction of oxygen in oxide, MO
P_1 = mole fraction of SiO_2 or B_2O_3 or P_2O_5
P_2 = mole fraction of metallic oxide, MO
K_1 and K_2 = constants whose values vary from one glass type to another (Table XXIV).

TABLE XXIV

VALUES OF K_1 AND K_2

Systems	K_1	K_2
SiO_2–PbO	4.34	5.23
SiO_2–Na_2O	4.698	4.934
B_2O_3–PbO	2.50	5.50
P_2O_5–PbO	3.70	3.70

A knowledge of ionic refractions for O^{Si}, O^{Si-M}, O^{M}, Si^{4+}, M^{x+}, and other ions and of molecular refractions for component oxides enabled Kordes to formulate equations for the calculation of molecular refractions for silicate-, borate-, and phosphate-glasses.

Equations (79) and (80) are given for silicate-glasses as follows: (i) From 0 to 66.7 mole percentage of MO,

$$K = P_1 \cdot K_{Si} + P_2 \cdot K_M + (2P_1 - P_2) \cdot K_{OSi} + 2P_2 \cdot K_{OSi-M}$$
$$= P_1 \cdot K_{SiO_2} - P_2 \cdot K_{OSi} + P_2 \cdot K_M + 2P_2 \cdot K_{OSi-M}. \quad (79)$$

(ii) From 66.7 to 100 mole percentage of MO,

$$K = P_1 \cdot K_{Si} + P_2 \cdot K_M + 4P_1 \cdot K_{OSi-M} + (P_2 - 2P_1) \cdot K_{OM}. \quad (80)$$

wherein K and K_X = respectively molecular and ionic refractions for glass and any component, X.
P_1 and P_2 = mole fractions of SiO_2 and MO, respectively.

The expressions for borate and phosphate glasses were similar in nature to these formulas.

In the system P_2O_5–PbO, a linear relationship existed for the composition range up to 61 mol percent of PbO. Kordes explained that oxygen introduced by PbO had the same effect on molecular refraction as the oxygen in P_2O_5 because the highly charged P ion attracted oxygen ions from the sphere of influence of Pb^{++}. In the system B_2O_3–Na_2O, oxygens introduced with Na_2O became linked directly to boron atoms, a fact which caused a triangular to tetrahedral change in coordination configuration. As no B–O linkages were broken, no change in molecular refraction occurred, and composition-refraction graphs for this series were linear. This finding was in agreement with that of Biscoe and Warren.[101(a)]

Similar methods of treatment were applied to polycomponent glasses arbitrarily converted into pseudobinary systems. Good agreement between calculated and experimental values was claimed. Ionic refractivities for the systems studied follow.

(*a*) $MgSiO_3$–$CaSiO_3$ (data after Peddle and Larsen),

Si	Ca	Mg	O^{Si}	O^{Si-Ca}	O^{Si-Mg}
0.1	1.40	0.26	3.673	4.50	4.02

(*b*) $NaAlSi_3O_8$–$CaAl_2Si_2O_8$ (data after Day and Allen, and Larsen),

Si	O^{Si}	Al	O^{Al}	Na	Ca
0.1	3.673	0.17	4.342	0.475	1.40

(*c*) $Pb_3(BO_3)_2$–Pb_2SiO_4 (data after Kordes),

Si	B	Pb	O^{Si-Pb}	O^{B-Pb}
0.1	0.05	9.13	4.94	4.75

It has been mentioned that Kordes calculated molecular volumes for glasses in the systems SiO_2–Na_2O, SiO_2–PbO, and P_2O_5–PbO from the ionic radii and molecular volumes of the components. In the discussion are presented his general considerations for the calculation of the molecular refractivities of silicate, borate, and phosphate glasses from ionic refractivities. By the familiar equation,

$$K = \frac{n^2 - 1}{n^2 + 2} \cdot V,$$

Kordes[112] was then able to calculate refractive indices in the system P_2O_5–PbO approximately within the limit of experimental error throughout the whole range of composition. He has thus succeeded in calculating the refractive index of a glass from a knowledge only of the ionic radii and ionic refractivities of its constituent ions and the molecular volumes of the pure components in that glass. Kordes' work is significant in that it suggests the possibility of future development in the calculation of physical constants from fundamental ionic units and reveals some indications as to the structure of glass.

Kordes[113] presented a new approach to this subject through his calculations of interionic distances from ionic refractivities for the glasses SiO_2 and B_2O_3. His results compare favorably with similar values determined by Biscoe and Warren[101(a)] and furnish additional information toward a better understanding of the physical concept of refractivity.

Geffcken[114] has pointed out an interesting similarity between aqueous solution and glass. In pure silica glass, the connection of each O atom with two different Si atoms may be represented by $(^1/_4Si) - O - (^1/_4Si)$. In the equation,[115]

$$(^1/_4Si) - O - (^1/_4Si) + Na_2O = 2 \cdot (^1/_4Si) - O^- + 2Na^+,$$

representing the addition of one mole of Na_2O, it will be noted that the "structure" or "frame" oxygen ions have changed to "anion" types. As Bernal and Fowler have indicated that the structure of water may be tetrahedral, the parallel expression for aqueous solution is

$$H - O - H + Na_2O = 2OH^- + 2Na^+.$$

[112] E. Kordes, "Physicochemical Investigation of Fine Structure of Glasses: II," *Z. physik. Chem.*, **B43**, 119–52 (1939); *Ceram. Abs.*, **19** [1] 10 (1940).

[113] E. Kordes, "Determination of Atomic Distances from Refraction," *Z. physik. Chem.*, **B44** [4] 249–60 (1939); *ibid.*, **B44** [5] 327–43 (1939).

[114] W. Geffcken, "Molecular Refraction in Glasses," *Glastech. Ber.*, **17** [8] 237–40 (1939); *Ceram. Abs.*, **19** [1] 9 (1940).

[115] (*a*) B. E. Warren and A. D. Loring, "X-Ray Diffraction Study of Structure of Soda-Silica Glass," *Jour. Amer. Ceram. Soc.*, **18** [9] 269–76 (1935).

(*b*) B. E. Warren and J. Biscoe, "Fourier Analysis of X-Ray Patterns of Soda-Silica Glass," *ibid.*, **21** [7] 259–65 (1938).

Some justification for this similarity is found in the fact that the ionic refractions of "frame" oxygen in water and in glass are 3.71 and 3.72_4, respectively, and that the respective refraction values of $(OH^- + Na^+)$ and $(RO^- + Na^+)$ are 5.30 and 5.17_3 to 5.40_9.

V. Discussion and Conclusions

The relation of density, refractive index, and specific refractivity to the composition of glass is dependent on the type of glass and on the composition range studied. Existing data and our present knowledge of the constitution of glass are not sufficient to justify the hope for a general relationship to fit all glass systems and all limits of composition. At the present time, only approximate, generalized formulas for wide ranges and accurate expressions for limited ranges of composition are possible. Equations of the first type have been investigated by Finn and his co-workers, who found that the specific refractions of forty-seven glass-forming oxides are linearly additive. Comprehensive studies of a similar nature are credited to Gilard and Dubrul and to Biltz, Weibke, and Schrader-Traeger. Expressions of the second type are more numerous and are usually accurate when applied to specific systems.

In any investigation, it is important to state the limitations of formulas or equations derived to express a certain relationship. For accurate work, compositions should be expressed in terms of chemical analyses, although the method of synthesis of Morey and Merwin[78] at times may be applicable. Because density and refractive index vary with the heat treatment of glass, modes of preparation that will yield reproducible results should be adopted.

From a consideration of modern concepts of glass constitution, it is logical to suggest that calculation factors should be so chosen as to represent the constituent ions. The use of constants derived for "component" oxides and compounds is to be avoided, although many authors have attributed unusual graphic results to the existence of compounds in glass.

Any factor or constant selected should produce mathematically accurate results and should possess physical rather than empirical significance. The failure of many authors to obtain linear relationships between physical properties and composition is understandable, as it is improbable that a single empirical factor should have been chosen to represent an entire composition range.

The reviewers suggest the adoption of the method of partial quantities advocated by Lewis and Randall[116] in their study of aqueous solutions. Accordingly, any property of a system may be represented or calculated by equation (81).

$$G = m_A \cdot \bar{G}_A + m_B \cdot \bar{G}_B + m_C \cdot \bar{G}_C + \ldots . \qquad (81)$$

wherein G = any property of a system, such as volume or refraction

$m_A, m_B, \ldots$ = moles or other weight-units of components A, B, etc.

$\bar{G}_A, \bar{G}_B, \ldots$ = their respective partial molal quantities or specific contributions of properties.

$\bar{G}$ values vary with concentration and may be complicated or simple functions of m; if $\bar{G}$ can be represented by a simple constant, linear additivity results.

Partial quantities may be useful in thermodynamic considerations of glass systems, although at the present time, the necessary data are lacking. In the calculation of interionic distances from ionic refractivities,[113] partial molal refractivities are essential factors. The usefulness of partial quantities as applied to the constitution of glass is illustrated. Finn and his associates found the partial specific volume of SiO_2 in soda-silica glasses to be 0.4540, 0.4371, and 0.4280 ml. per gram, respectively, for the ranges 100 to 75%, 75 to 63%, and 63 to 50% of SiO_2. It is interesting to note that between the limits of 75 to 100% of SiO_2, the volume of SiO_2 is identical with that of vitreous SiO_2 and is not affected by the presence of Na_2O. The interionic distances for Si and O become progressively smaller with increase in Na_2O content, but they have constant values for each range specified. The ratio of interionic distances for adjacent composition ranges is equal to the ratio of the cube root of the respective partial specific volumes. Similar reasoning applies to the case of Na_2O whose partial specific volume is 0.2830, 0.3327, and 0.3486 ml. per gram, respectively, for the ranges already indicated. The reviewers were able to calculate these interionic distances (Table XXV), as the values for the bond distances Si–O and O–O in vitreous silica are 1.62 Å and 2.65 Å, respectively,[117] and that for Na–O in soda-silica glass (64 to 80% SiO_2) is 2.35 Å.[116(b)]

The calculation of partial quantities for aqueous solutions is usually limited to binary systems. The reviewers have extended method II of Lewis and Randall[116] to ternary and polysystems. It was found applicable for the computation of partial quantities in proposed glasses whose compositions would undergo a regular variation. For certain existing data, a modification of the procedure indicated that similar calculations could be made.

[116] G. N. Lewis and M. Randall, Thermodynamics, chapters IV and VIII. McGraw-Hill Book Co., New York, N. Y., 1923.

[117] B. E. Warren, H. Krutter, and O. Morningstar, "Fourier Analysis of X-Ray Patterns of Vitreous SiO_2 and B_2O_3," *Jour. Amer. Ceram. Soc.*, **19** [7] 202–206 (1936).

TABLE XXV

CALCULATED INTERIONIC DISTANCES IN SODA-SILICA GLASS AFTER THE REVIEWERS

Ranges SiO_2 (%)	SiO_2					Na_2O		
		Si–O distance (Å)		O–O distance (Å)			Na–O distance (Å)	
	Partial sp. vol.	Obs.	Calc.	Obs.	Calc.	Partial sp. vol.	Obs.	Calc.
50–63	0.4280		1.59		2.60	0.3486		2.39
63–75	0.4371		1.60		2.62	0.3327	2.35	
75–100	0.4540	1.62		2.65		0.2830		2.23

Glasses whose compositions varied irregularly required the formulation of an expression containing high exponential values of m_A, m_B, ... Composition-property data for all glasses in a series may be substituted in this equation, and the resulting expression may be solved simultaneously to obtain the desired constants.

The plot of a property, G, on the third dimension above a triangular composition diagram will produce a curved surface. Method IV of Lewis and Randall[116] suggests that the intercepts of a tangent plane of any point of this surface with the three vertical axes of the apices will represent the partial quantities of the three components at that point. The situation is simplified when such a plot produces plane rather than curved surfaces.

Although this review has been restricted to a discussion of density, refractive index, and specific refractivity, the accompanying literature search has revealed that other properties of glass may possibly be treated in a similar manner. With the present limited knowledge of the structure of glass, it has been possible to calculate density from interionic distances and the latter in turn from ionic refractivities. A better understanding of the constitution of glass may completely change existing methods of treatment of property-composition relations. It is hoped that the ideas contained herein may serve to stimulate future investigation.

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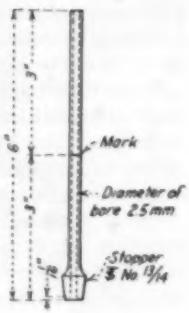
SPECIAL STOPPER FOR MERCURY VOLUMETER*

Editor's Note: The accompanying cut and description of the stopper for the Seil volumeter was received too late to accompany the publication of the paper, "Apparatus and Method for Determination of Porosity in Refractory Materials," by Gilbert E. Seil, Bradford S. Tucker, and H. A. Heiligman. This paper was published in the November issue of *The Journal*, pp. 330–33.

Stopper for Volumeter

The stopper has proved to be advantageous when accurate readings on the volumeter are desired. The glass stopper is inserted after the mercury has risen to the neck in the separatory funnel and the air has been released by stirring. The reading is taken when the mercury reaches the mark on the glass stopper instead of when it reaches the projection, I, of the steel cap, E, as described in the original article.

An added footnote reference is H. H. Macey and F. H. Clews, "Measurement of Apparent Porosity," *Trans. Brit. Ceram. Soc.*, **39** [9] 279–88 (1940).



* Received October 12, 1940.

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Compiled by

THE AMERICAN CERAMIC SOCIETY

ROSS C. PURDY, *Editor*
EMILY C. VAN SCHOICK, *Assistant Editor*
VIRGINIA BENEDICT, *Assistant*

Vol. 23 December, 1940 No. 12

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The reference number in parentheses refers to the *monthly* issue of the *Journal;* the number following is the *page* number.

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JOURNAL

of the

American Ceramic Society

A Monthly Journal Devoted to the
Arts and Sciences Related to
the Silicate Industries

Volume 23, 1940

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Compiled by

THE AMERICAN CERAMIC SOCIETY

ROSS C. PURDY, *Editor*

Vol. 19 December, 1940 No. 12

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Compiled by

The American Ceramic Society

Volume 19, 1940

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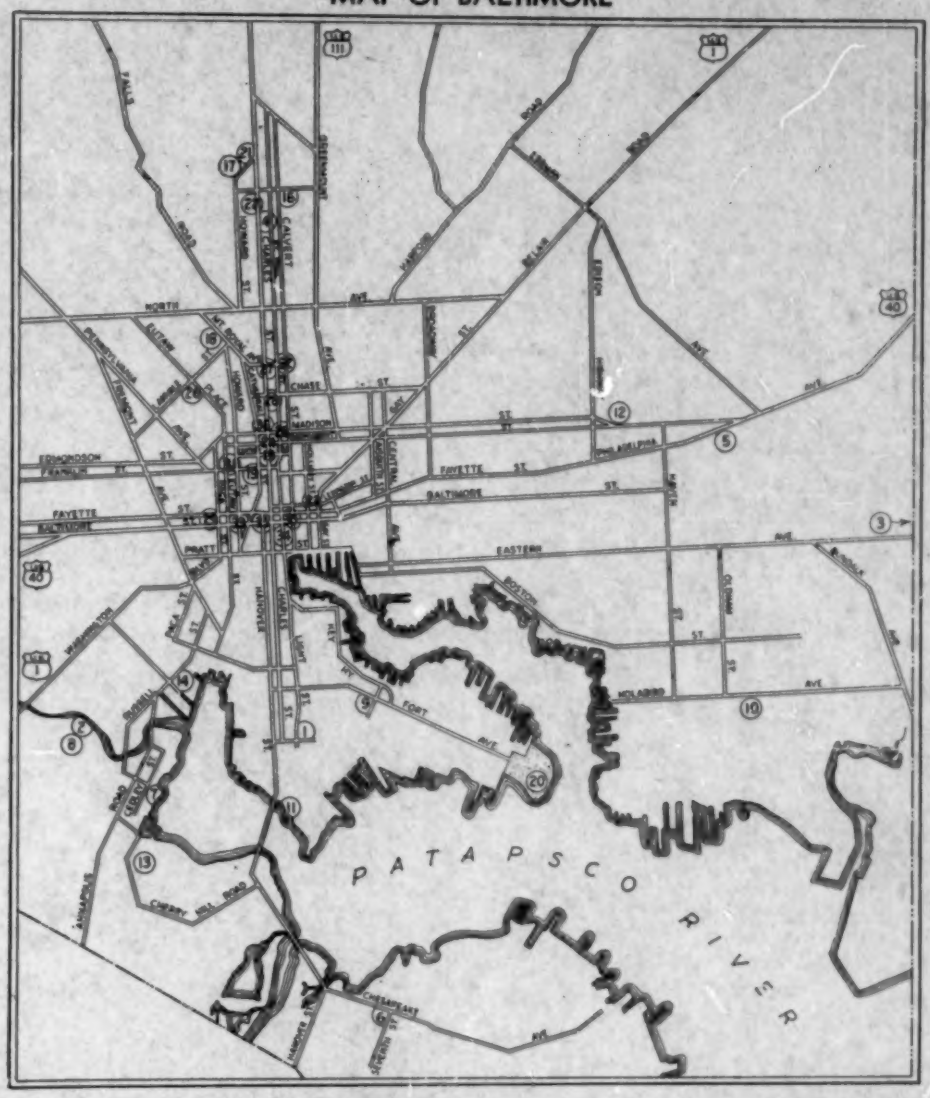
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FORTY-THIRD ANNUAL MEETING, AMERICAN CERAMIC SOCIETY

BALTIMORE, MARYLAND, MARCH 30 to APRIL 5, 1941

BALTIMORE HOTELS

The true test of a meeting city is not only the number of its hotels but their quality. Baltimore, with a tradition of hospitality founded on good food and good service, enjoys an established reputation in accommodating visitors to the city and making them feel at home. A reputation for the enjoyment of living has made Baltimore a haven for travelers. Baltimore has several consistently good hotels. The larger hotels are all located within a radius of a few blocks.

If you have ever been a visitor to Baltimore, you probably will recall with pleasure one of the following, a few of Baltimore's sixty hotels:

Lord Baltimore	Mt. Royal
Emerson	New Howard
Southern	Arundel
Belvedere	Stafford
Rennert	Congress

Altamont

ENTERTAINMENT AND RECREATION IN BALTIMORE

With (1) its rich historical background, (2) diversified industrial and world-wide commercial activity, (3) cultural and educational attainments, and (4) proximity to the Nation's Capitol, to picturesque and colonial Annapolis, home of the U. S. Naval Academy, and the near-by Chesapeake Bay, aptly described as the "Little Mediterranean of America," offering exceptional and almost unlimited post-Meeting recreational opportunities, Baltimore is truly one of America's most interesting, complete, and diversified meeting cities.

BALTIMORE, MARYLAND, CLIMATE

Baltimore's temperate climatic conditions are favorable for meetings at any time of the year as shown by the following mean temperatures based on figures compiled by the United States Weather Bureau over a long period of time:

Winter	35.5 degrees	Spring	53.4 degrees
Summer	75.1 degrees	Fall	57.7 degrees